

## Reentrant Peak Effect in an anisotropic superconductor 2H-NbSe<sub>2</sub>: Role of disorder

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The reentrant nature of the **Peak Effect (PE)** is established in a single crystal of anisotropic superconductor 2H-NbSe<sub>2</sub> via electrical transport and dc magnetization studies. The role of disorder on the reentrant branch of the **PE** has been examined in three single crystals with varying levels of quenched random disorder. Increasing disorder presumably shrinks the (H,T) parameter space over which vortex array retains spatial order. Although, the upper branch of the **PE** curve is somewhat robust, the lower reentrant branch of the same curve is strongly affected by disorder.

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A magnetic flux line lattice (FLL) or vortex lattice (VL) with lattice constant  $a_0 \propto \frac{1}{\sqrt{Field}}$  was predicted [1] in the entire mixed state of a type-II superconductor within a mean field model. The advent of high temperature superconductors (HTSC) focused attention on the influence of thermal fluctuations on VL and led to the prediction [2] of a Vortex Liquid State with an unusual reentrant phase boundary in the magnetic field-temperature (H, T) plane such that at a fixed T, the melting phase boundary (H<sub>m</sub>(T)) is encountered *twice* upon increasing H [3]. A dilute vortex liquid phase is expected to form above the lower critical field H<sub>c1</sub>, where a<sub>0</sub> ≥ λ, the magnetic penetration depth λ gives a measure of the range of repulsive interaction that governs the FLL. A highly dense vortex liquid phase is expected below the upper critical field, where a<sub>0</sub> ≥ 2ξ; ξ is the coherence length, i.e., the radius of the vortex core. The two branches comprising reentrant melting phase boundary join around the so-called “nose” temperature above which the vortex solid phase is thermally melted at all field values. The upper branch of the melting curve is given approximately by eqn. 5.5 of Ref.3,

$$B_m = \beta_m \left( \frac{c_L^4}{G_i} \right) H_{c2}(0) \left( 1 - \frac{T}{T_c} \right)^2, \quad (1)$$

where c<sub>L</sub> is the Lindemann parameter, G<sub>i</sub> is the Ginzburg number (= (1/2)(k<sub>B</sub>T<sub>c</sub> / H<sub>c</sub><sup>2</sup>ξ<sup>3</sup>ε/8π)<sup>1/2</sup>) and β<sub>m</sub> ≈ 5.6. G<sub>i</sub> is much larger in the Cuprate HTSC systems as compared to that in low T<sub>c</sub> alloys, which facilitates the observation of the dense vortex liquid phase in HTSC

systems [3]. However an experimental observation of the dilute vortex liquid phase, has remained elusive. On the lower branch, the melting phase boundary is governed by eqn. 5.19 of Ref.3,

$$\frac{\lambda}{a_0} e^{\left( \frac{a_0}{\lambda} \right)} = \beta_m' \left( \frac{c_L^4 \kappa \ln \kappa}{G_i} \right) \left( \frac{T_c}{T} \right)^2 \left( 1 - \frac{T}{T_c} \right)^2 \quad (2)$$

where a<sub>0</sub>=[2/3<sup>1/2</sup>(ϕ<sub>0</sub>/B)]<sup>1/2</sup> and β<sub>m</sub>'=0.5. Solving eqn.(2) gives B<sub>m</sub>(T→0)=H<sub>c1</sub>(0)[ln(T/T<sub>c</sub>)]<sup>-2</sup>. The large value of κ in the Cuprates presumably restricts the dilute vortex liquid phase to very small induction values, making its experimental observation a challenging task.

A further difficulty in the observation of the vortex liquid phases arises due to the ubiquitous quenched disorder (i.e., pinning centers) in all real systems which adversely affects the stability of the ordered vortex solid phase. The effect of disorder is expected to be especially strong for dilute vortex arrays and may mask the observation of an underlying dilute liquid - dilute solid melting transition [4,5].

In the context of quenched disorder, the phenomenon of the peak effect (PE), an anomalous increase in the critical current, is continuing to receive a great deal of attention as it marks the competition between interaction and disorder (i.e., elastic energy versus pinning energy) [6–13]. In a collective pinning description [14] of superconductors, J<sub>c</sub> relates inversely to the volume V<sub>c</sub> of Larkin domain (J<sub>c</sub> ∝  $\frac{1}{\sqrt{V_c}}$ ) over which FLL is correlated. The anomalous peaking in J<sub>c</sub> indicates a rapid decrease

in  $V_c$  due to softening of elastic moduli of FLL at the incipient melting transition [6,11,12,14], thus making the PE yet another signature of the loss of order of the vortex array. The identification of PE as a “phase boundary” remains among the more complex and controversial questions in type II superconductors in general [6–12].

Ghosh *et al* [10] showed that in a weakly pinned vortex lattice of hexagonal 2H-NbSe<sub>2</sub> ( $T_c \approx 7.1$  K), the **PE** curve, which is the locus of peak temperatures  $T_p(H)$  (determined from the magnetic shielding response, i.e., ac susceptibility  $\chi'$  measurements), bore a striking resemblance to the theoretically proposed reentrant [2,3] melting phase boundary  $T_m(H)$ . The *turnaround* of reentrant  $T_p(H)$  curve has been reported [10] to occur when  $a_0$  (of  $4000$  Å<sup>0</sup> at  $H \approx 150$  Oe)  $\approx \lambda_c$  (in 2H-NbSe<sub>2</sub>), and it is followed by a rapid broadening of **PE**. The **PE** ultimately became undetectable below  $30$  Oe [10]. Since PE is directly related to pinning, its variation with varying pinning strength is crucial in providing an understanding of the complex interplay between thermal and quenched disorder in destabilizing the ordered phase. In this letter, we report new results on the reentrant nature of PE curve and the effect of quenched random disorder on it via electrical transport measurements and an equilibrium dc magnetization experiment on a specimen of 2H-NbSe<sub>2</sub> utilized by Ghosh *et al* [10] and magnetic shielding ( $\chi'$ ) response studies on two different crystals of the same compound with qualitatively different levels of quenched random disorder. The conventional dc electrical transport results not only fortify the earlier claim [10] of reentrant nature of the  $T_p(H)$  curve but also corroborate the previous finding that **PE** broadens across fields where *turnaround* in  $T_p(H)$  takes place. The magnetization hysteresis data also confirm the double crossover of  $T_p(H)$  curve in an isothermal scan near the *turnaround* region. We find that though the upper (higher  $H$ ) branch of  $T_p(H)$  curve is somewhat robust, the lower reentrant branch is strongly influenced by disorder. Decreasing pinning strength results in the ‘nose’ region being pushed down to fields lower than that reported earlier [10], whereas increasing pinning could make the ‘nose’ feature completely disappear. These findings provide quantitative estimate on how increasing disorder shrinks the ( $H, T$ ) space over which vortex array retains spatial order.

2H-NbSe<sub>2</sub> has an appreciable value ( $\approx 3 \times 10^{-4}$ ) of  $G_i$ , which lies between those of HTSC and conventional superconducting alloys [11]. The weak pinning characteristic of single crystals of this compound [ $J_c/J_0 \sim 10^{-5}$  to  $10^{-7}$ , where  $J_c$  is the critical current density and  $J_0$  ( $\approx 10^8$  A/cm<sup>2</sup>) is the theoretical depairing current density] facilitates the formation of VL in them [11] and easy variability of spatial correlation lengths in its specimen make them convenient test beds to observe phenomena which are a consequence of competition and interplay between interaction, thermal fluctuations and quenched random disorder. Three single crystals with increasing quenched disorder used in the present study comprise platelets be-

longing to the same batches as the specimen **X** used by Higgins and Bhattacharya [11], as the specimen **Y** used by Ghosh *et al* [10] and as the specimen **Z** used by Henderson *et al* [13]. The dc resistance data to ascertain **PE** temperatures in specimen **Y** were recorded following Ref. 11, the isothermal magnetization hysteresis data to identify the manifestation of **PE** in a similar sample were obtained on a Quantum Design Squid Inc. magnetometer following the *half scan technique* of Ravikumar *et al* [15] and ac susceptibility data in all three samples were recorded using a high sensitivity ac susceptometer [16]. The essential new findings are summarized in Figs. 1 to 3. All data presented are for  $H \parallel c$ , however, **PE** is observed in all orientations in 2H-NbSe<sub>2</sub> and the anisotropic Ginzburg-Landau description applies to this system [11].

Fig.1 shows the temperature variation of resistance,  $R$  vs reduced temperature ( $t = T/T_c(0)$ ), in the crystal **Y** at some of the low fields (120 Oe to 380 Oe) to locate the signature of the reentrant behavior in  $T_p(H)$  data. The inset (a) of Fig.1 shows that the peak in  $J_c$  at a given  $H$  manifests as an anomalous dip in  $R(t)$ . As emphasized in Ref. 11, the location of each  $T_p(H)$  is robust. The main panel of Fig. 1 shows portions of  $R(t)$  curves at different  $H$  and focusses attention onto the identification of  $t_p(H)$  values. The inset panel (b) of Fig.1 depicts the PE curve which passes through the  $t_p(H)$  values for  $120 \leq H \leq 380$  Oe. The shape of the PE curve clearly demonstrates its reentrant characteristics, with *turnaround* occurring at about 180 Oe, in excellent agreement with earlier results [10] obtained via  $\chi'(T)$  measurements. The **PE** behavior in a given  $R(t)$  curve starts at a temperature,  $T_{pl}$ , where the resistance starts to decrease (see  $t_{pl}$  mark for  $H=360$  Oe curve). The data in the main panel of Fig. 1 imply that significant broadening of **PE** occurs as  $H$  values are decreased below 200 Oe, and eventually below 120 Oe, the **PE** becomes so shallow that  $t_p(H)$  cannot be located precisely for  $H < 100$  Oe.

A further confirmation of the reentrant characteristic of **PE** curve (in sample **Y** with  $T_c(0) = 7.17$  K) is provided by the results of dc magnetization hysteresis measurements, an example of which is shown in Fig. 2. In an isothermal scan at  $T = 6.95$  K, we expect to encounter both the lower and upper branches of the reentrant **PE** curve. The inset panel of Fig. 2 shows a portion of the M-H hysteresis data at  $T = 6.95$  K. The **PE** on the upper branch manifests as an anomalous opening of the hysteresis loop before reaching the  $H_{c2}$  value. According to Bean’s Critical State Model [17], magnetization hysteresis  $\Delta M(H)$  (difference between forward and reverse magnetization values at given  $H$ ) is a measure of  $J_c(H)$ . To locate the existence of **PE** on the lower branch and to elucidate that the peak field  $H_p$  on the lower branch of **PE** curve **increases** as  $T$  **increases**, we show in Fig. 2 the variation of  $4\pi\Delta M$  ( $\propto J_c(H)$ ) vs  $H$  at 6.9 K, 6.95 K and 7.0 K in the low field region. It is to be noted that for  $H < 60$  Oe,  $\Delta M(H)$  values at 6.95 K are **lower** than those at 6.9 K and this reflects the normal behavior in  $J_c(H, T)$ . However, a crossover in  $\Delta M(H)$  curves occurs at about

70 Oe such that  $\Delta M(H)$  values at 6.95 K **exceed** those at 6.9 K, thereby exemplifying the observation of anomalous behavior [18] in  $J_c(H,T)$  due to the presence of **PE** on the lower branch at 6.95 K. It may be further noted that  $\Delta M(H)$  values at 7.0 K become larger than those at 6.95 K above 130 Oe. Thus, at 7.0 K the (lower) peak effect is seen to be encountered at a field value larger than that at 6.95 K, this establishes the central characteristic (i.e.,  $dT_p/dH > 0$ ) of the lower branch of the PE curve.

Fig. 3 shows the temperature variation of the in-phase ac susceptibility ( $\chi'$  vs reduced temperature  $t$ ) at a frequency of 211 Hz and in  $h_{ac}$  of 1 Oe(r.m.s.) at some of the chosen dc fields  $H_{dc}$  (0 to 2 kOe) in three crystals **X**, **Y** and **Z** of 2H-NbSe<sub>2</sub>. These crystals were grown by the same vapor transport method [19], but with starting materials of different purity such that the specimen **X** is the cleanest among the three and the specimen **Z**, the most disordered. The relative purity of the three crystals is reflected in the *width* of the superconducting transition in  $\chi'(t)$  response in zero field, which can be seen to progressively enhance in Figs. 3(a) to 3(c). In a  $\chi'(t)$  measurement at a given  $H$ , the **PE** is identified by an anomalous negative peak [9,10]. The peak temperatures  $T_p(H)$  are independent of frequency and amplitude of ac field  $h_{ac}$  in the parametric range of our experiment [20]. Fig. 3(a) shows that in the most weakly pinned crystal **X**, the **PE** peaks are very sharp; *their half widths are smaller than the width of superconducting transition in zero field*. This fact supports the association of  $T_p(H)$  with a possible phase transition, like, the phenomenon of change in spatial order of the vortex array across  $T_p$ .

Fig. 3(b) is based on the  $\chi'(t)$  response in crystal **Y** [10].  $T_p(H)$  values for  $H=300$  Oe and  $H=100$  Oe can be seen to be nearly the same, consistent with the reentrant nature of **PE** curve in crystal **Y** [10]. However, the **PE** in  $H=200$  Oe (data not shown) is significantly broader than that at  $H=300$  Oe. Fig. 3(c) shows the  $\chi'(t)$  response in the most strongly pinned [13] crystal **Z**. In this sample, the **PE** peak is considerably broad even at  $H=1000$  Oe as compared to the corresponding peaks in crystals **X** and **Y** (cf. Figs. 3(a) to 3(c)). The **PE** peak is barely discernible at  $H=300$  Oe in crystal **Z** and below 200 Oe, the **PE** manifests only as a hump/kink in  $\chi'(t)$  curve.

Fig.4 summarizes the  $T_p(H)$  data in three crystals **X**, **Y** and **Z** as  $H$  vs  $T_p/T_c(0)$  curves. For the sake of convenience and reference, we have also included in this figure the  $H_{c1}(t)$  and  $H_{c2}(t)$  curves obtained in crystal **X** [6]. Thus, from Fig.3 and Fig.4, it is apparent that enhanced quenched random disorder (pinning) results in the following effects :

- (i) For a given  $H$  ( $H \geq 300$  Oe), the **PE** peak occurs at a lower value of  $t$  (see, for instance, arrows marking  $T_p(H)$  values in three crystals at  $H=300$  Oe in Figs.3(a) to 3(c)). This amounts to a “lowering” of the upper branch of **PE** curve with enhanced pinning (see Fig.4). Also note that the **PE** broadens as the disorder increases.
- (ii) The reentrant lower branch of **PE** curve is clearly evident only for crystal **Y** with intermediate level of disorder

and not so for the other two crystals. The **PE** curve in crystal **X** shows a steep fall for  $200 < H < 30$  Oe and its turnaround characteristic presumably lies much below 30 Oe; however, **PE** in  $\chi'(T)$  data is unobservable in crystal **X** for  $H \leq 20$  Oe. In sample **Z**, the **PE** becomes so broad at 300 Oe that the precise location of  $T_p(H)$  below this field becomes somewhat ambiguous (see Fig.3(c)). However, it can be safely surmised that **PE** curve at lower fields ( $H < 500$  Oe) moves away from the values that can be extrapolated from  $T_p(H)$  vs  $H$  data at higher fields ( $H > 500$  Oe). Thus the effect of enhanced disorder (**X** to **Z**) is seen to result in “raising” to higher fields, the lower reentrant branch of the PE curve (in contrast to the upper branch).

The results of Figs. 3 and 4 provide the following general conclusions:

- (i) For the upper branch (i.e.,  $dT_p/dH < 0$ ) of the PE curve, increasing pinning reduces the volume of Larkin domain of FLL, which then requires less thermal fluctuations to melt/amorphize it around  $t_p$ . The quenched disorder and thermal fluctuations conspire together to destabilize the ordered phase and a “lowering” of the  $t_p(H)$  curve is seen.
- (ii) For the lower branch (i.e.,  $dT_p/dH > 0$ ), increasing pinning reduces the volume of Larkin domain as before. Thus, one needs enhanced interaction (i.e., increase in  $H$ ) to stabilize the ordered phase and the “raising” of the PE curve occurs.
- (iii) Increasing pinning has the general effect of masking the difference between a vortex solid and a vortex liquid, converting both into a “glassy” state. A clear distinction between the two phases is no longer possible, the transformation process is more gradual and thus a broadening of the PE results.
- (iv) As can be ascertained from the main panel of Fig.1, the onset (at  $t_{pl}$ ) of PE, which is apparently driven by quenched disorder, is also reentrant, just as the vortex melting transition is expected to be. Recent muon spin rotation studies [21] on crystals of 2H-NbSe<sub>2</sub> have provided microscopic evidence in favour of a sudden change in spatial order of FLL at the onset of PE, i.e., at  $T_{pl}$ .

Finally, it is of interest to examine the scenario emerging from Fig.4 with the expectations of theoretical studies in the ( $H$ ,  $T$ ) region of our experiments. In the cleanest crystal **X**, where we observe only the upper branch of **PE** curve, which most closely marks the transformation of the ordered solid into the *pinned “liquid” state* and a narrow region separates this line from  $T_c(H)$  line (cf. Figs. 6, 24 and 25 of Ref. 3 and our Fig.4). In between  $T_p(H)/H_p(T)$  and  $T_c(H)/H_{c2}(T)$  lines, the pinned liquid becomes unpinned at  $H_{irr}(T)$  (see Fig. 2(a) for location of  $H_{irr}$  at 6.95 K). Fitting the higher field ( $H > 200$  Oe) branch of **PE** curve to a standard quadratic Linde-mann relationship (see eqn. (1)) with  $G_i \sim 3 \times 10^{-4}$  and  $H_{c2}(0) \approx 4.6$  T [6], we get a reasonable value for Linde-mann number  $c_L = 0.15$ . A pronounced departure (in  $T_p(H)$  at  $H \leq 200$  Oe) away from the upper portion appears in accord with a recent theoretical work of Blatter

and Geshkenbein [22], who found a similar rapid drop in FLL melting curve (in the absence of pinning effects), away from the  $H_{c2}$  phase boundary, at low fields (i.e., above the *turnaround* feature of the melting boundary). The reentrant lower branch of **PE** curve may thus be at still lower fields in crystal **X**, as proposed theoretically [22], and outside the detection capability of the present measurements. In crystal **Y**, with intermediate level of disorder, Ghosh *et al* [10] estimated  $c_L$  as about 0.17 from higher  $H$  portion of the  $T_p(H)$  data. They also showed that if the Nelson-LeDoussal line [4] indeed marks the crossover from an interaction dominated to a disorder-dominated region, then the ratio of entanglement length  $L_E$  to the pinning length  $L_c$  becomes approximately equal to 1 ( $L_c/L_E \sim 1$ ) at  $B=30$  Oe and  $t=0.975$ .  $L_c/L_E$  is given as [3],

$$\frac{L_c}{L_E} = \left( \frac{\pi \kappa^2 \ln(\kappa)}{\sqrt{2}} \right) \left( \frac{a_0^2}{2\pi \lambda(0)} \right) \frac{(J_c)^{1/2}}{(G_i j_o)^{1/2}} \frac{(1-t)^{4/3}}{t}, \quad (1)$$

where various symbols have their usual meaning (see eqn. 6.47 of Ref.3). Below 30 Oe, the vortex array would be in disentangled liquid or glass state [3-5]. Eqn.1 implies that the field at which crossover to glassy state can occur is  $\propto J_c^{1/2}$ .  $J_c$  is larger in crystal **Z** by nearly a factor of 50 as compared to that in crystal **Y**. This, coupled with the observation that in crystal **Z** the **PE** becomes difficult to discern below  $t \approx 0.96$ , yields a value of crossover field of  $\sim 400$  Oe in sample **Z**, which demonstrates a very reasonable agreement with an observed value of 300 Oe in our experiment.

In summary, we have demonstrated the variation of the PE at high temperatures and low fields, and especially of its lower reentrant, branch is experimentally measurable. The results show not only the shifts in the relevant transitions/crossover lines separating the ordered phase from the disordered ones, they also illustrate how the distinction between them is blurred as quenched disorder is varied. Some aspects of the results could be semi-quantitatively explained by the available theories [4,5,22,23]. Nevertheless, a more detailed theoretical analysis of the low field - high temperature regime, especially on the liquid - dilute solid transformation process, is necessary.

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FIG. 1. The inset panel (a) shows the temperature variation of Resistance ( $R$  vs reduced temperature,  $t=T/T_c(0)$ ) in  $H_{dc}=280$  Oe ( $\parallel c$ ) in the crystal **Y** of 2H-NbSe<sub>2</sub>. The Peak Effect (PE) region can be identified by an anomalous dip and peak temperature  $t_p$  is marked. The main panel shows portions of  $R$  vs  $t$  curves in the PE regions at some chosen dc fields in the range  $140$  Oe  $\leq H \leq 360$  Oe. The marked  $t_p(H)$  values elucidate that  $t_p(H)$  increases as  $H$  decreases ( $dH/dt_p < 0$ ) from 360 Oe to 200 Oe and thereafter (for  $H < 200$  Oe)  $dH/dt_p > 0$ . The inset panel (b) focusses attention onto the reentrant nature of locus of  $t_p(H)$ .

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FIG. 2. Isothermal magnetization hysteresis data [ $4\pi\Delta M = 4\pi(M_R - M_F)$ ] for  $H_{dc}\parallel c$  in the crystal Y of 2H-NbSe<sub>2</sub> at the temperatures indicated. The arrows identify the fields corresponding to onset of PE phenomenon on the lower (reentrant) branch of PE curve at temperatures of 6.95 K and 7.0 K. It is to be noted that the field at which (lower) PE occurs at 7.0 K is larger than that at 6.95 K. The inset panel shows a portion of the M-H loop showing upper PE at 6.95 K. (Upper) Peak field  $H_p$  and irreversibility field  $H_{irr}$  at 6.95 K have been identified.

FIG. 3. In phase AC ( $f=211$  Hz,  $h_{ac} = 1$  Oe (r.m.s.)) susceptibility ( $\chi'$ ) vs reduced temperature ( $T/T_c(0)$ ) in crystals **X**, **Y** and **Z** of 2H-NbSe<sub>2</sub> in fixed dc fields ( $\parallel c$ ) as indicated. The arrows mark the  $t_p$  values at  $H_{dc}=300$  Oe in different crystals.

FIG. 4. Magnetic phase diagram in three crystals of 2H-NbSe<sub>2</sub>. PE curves and  $H_{c2}(T)$  curve correspond to  $T_p(H)/T_c(0)$  and  $T_c(H)/T_c(0)$  values obtained from  $\chi'(t)$  data as in Fig.3. For crystal Y, the two open squares lying on the upper and lower branches of PE curve identify the peak field ( $H_p$ ) and crossover field at 6.95 K as in Fig.2.

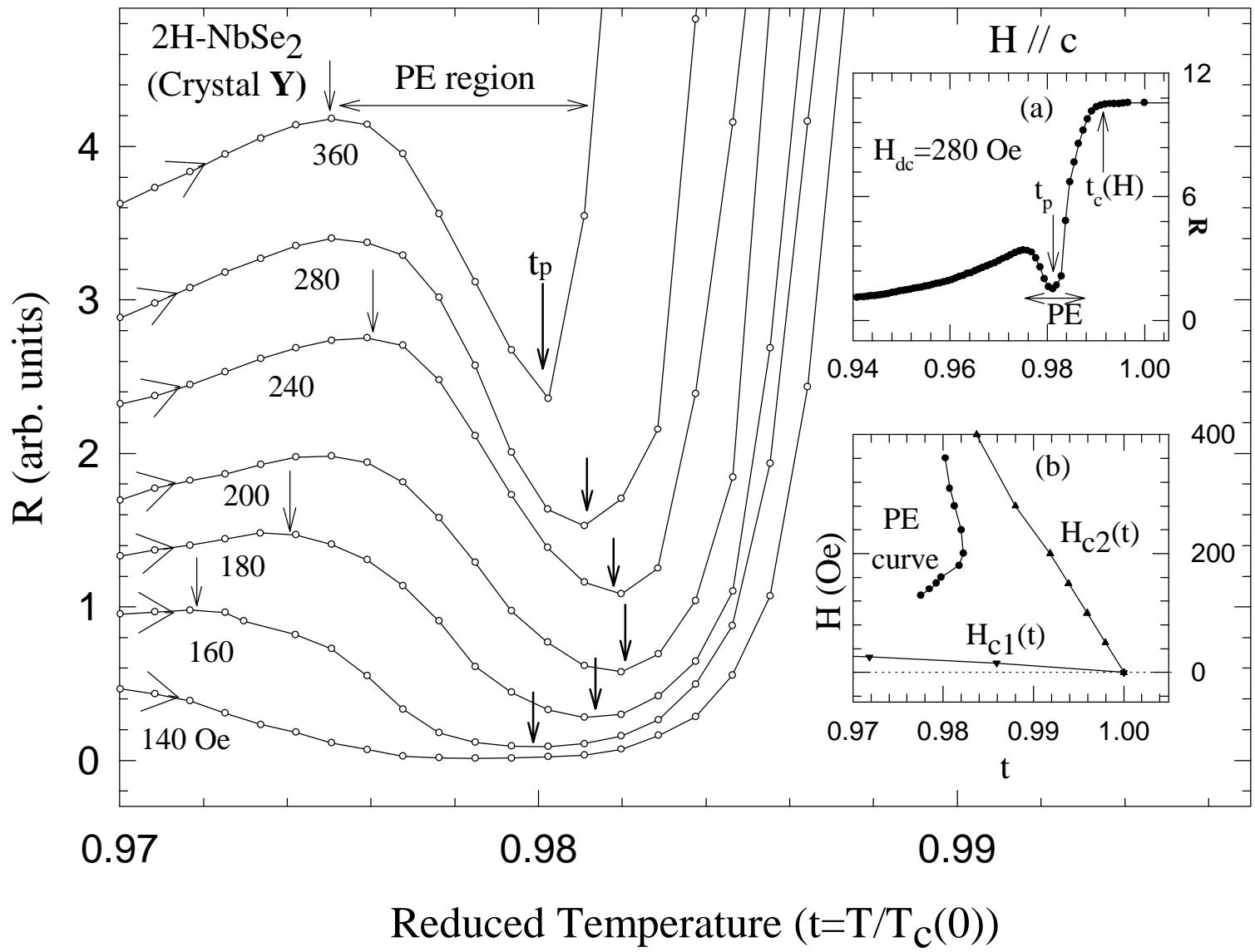


Fig.1 (S. S. Banerjee et al)

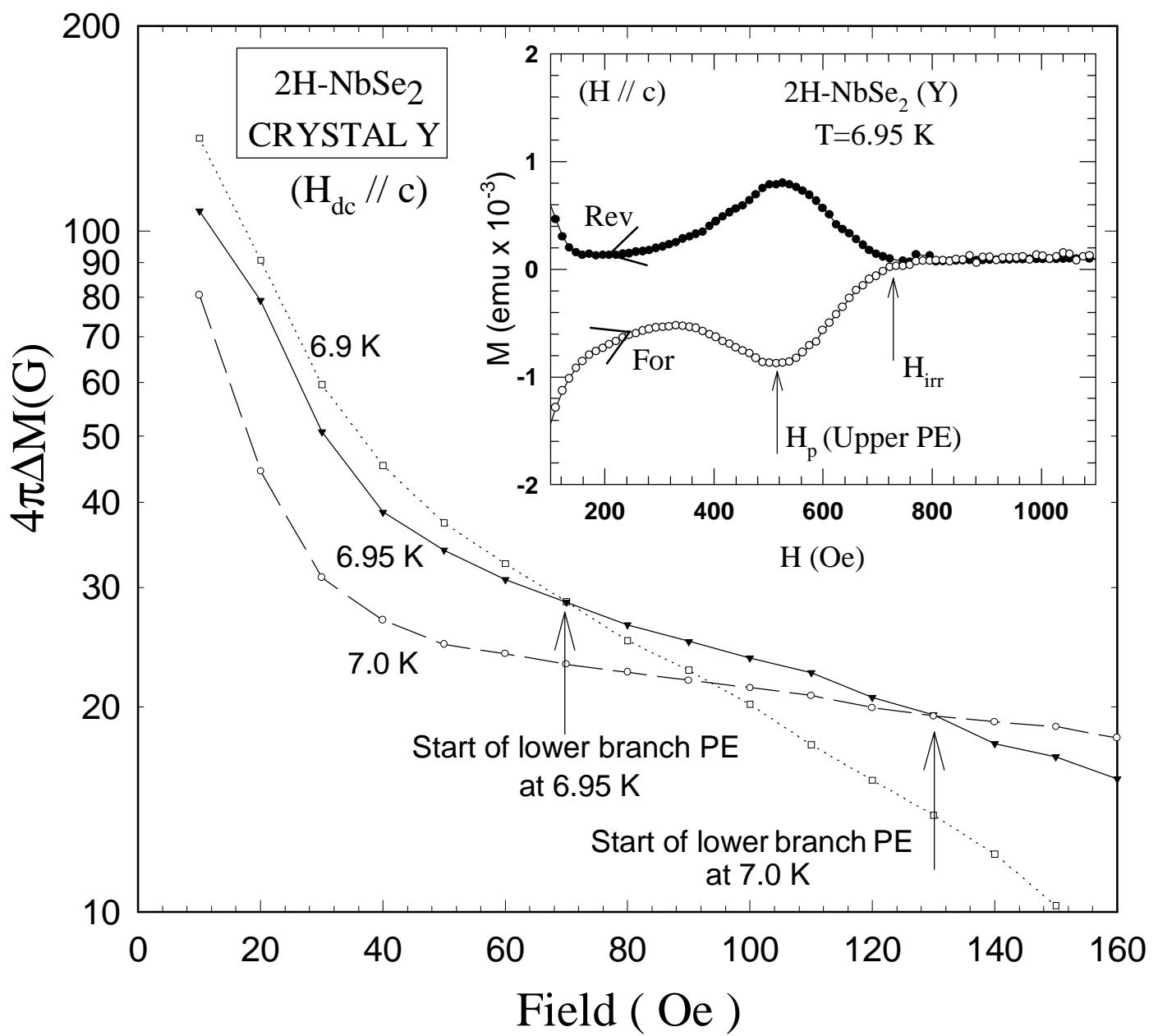


Fig. 2 (S. S. Banerjee et al)

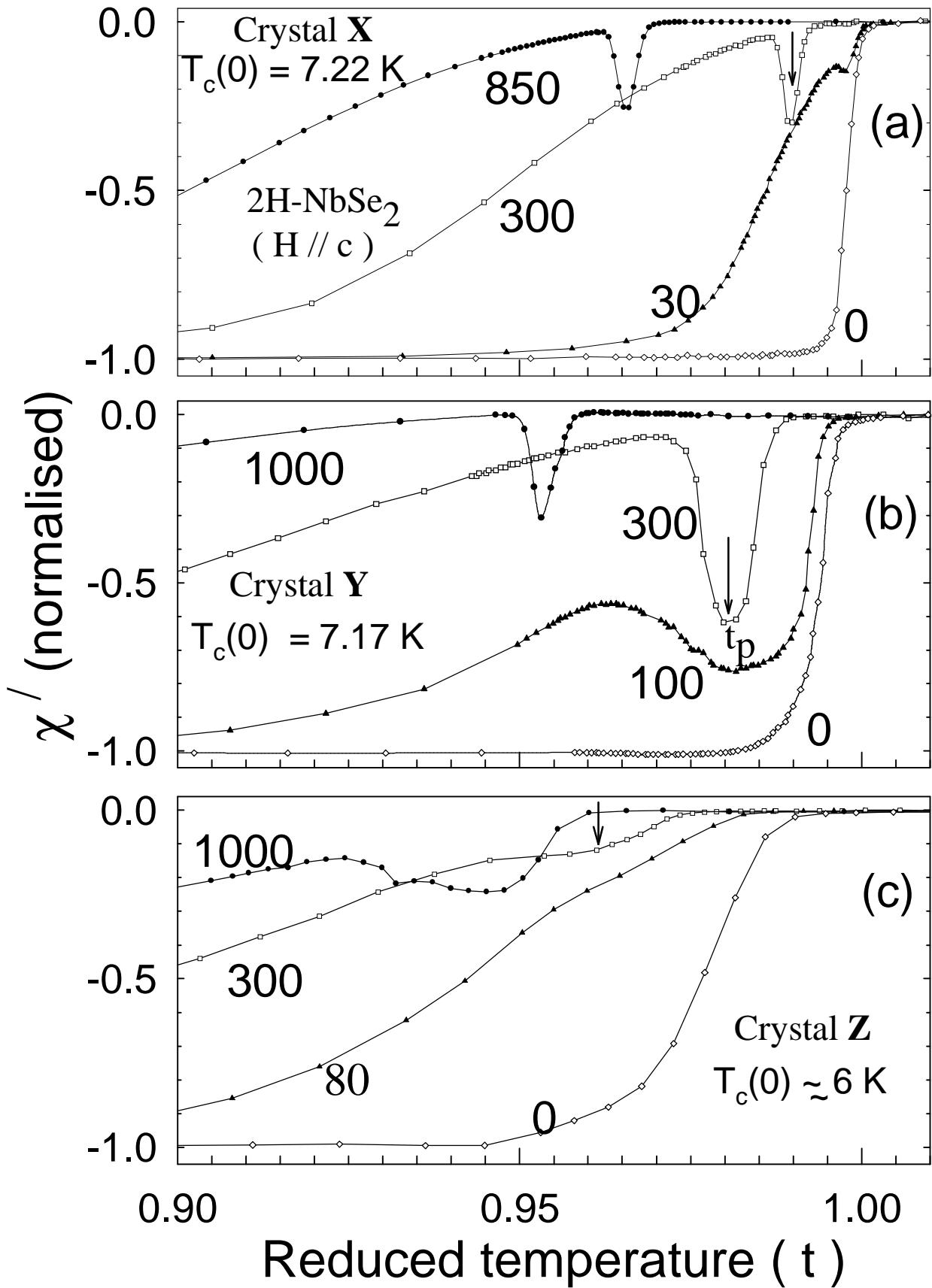


Fig.3 (S. S. Banerjee)

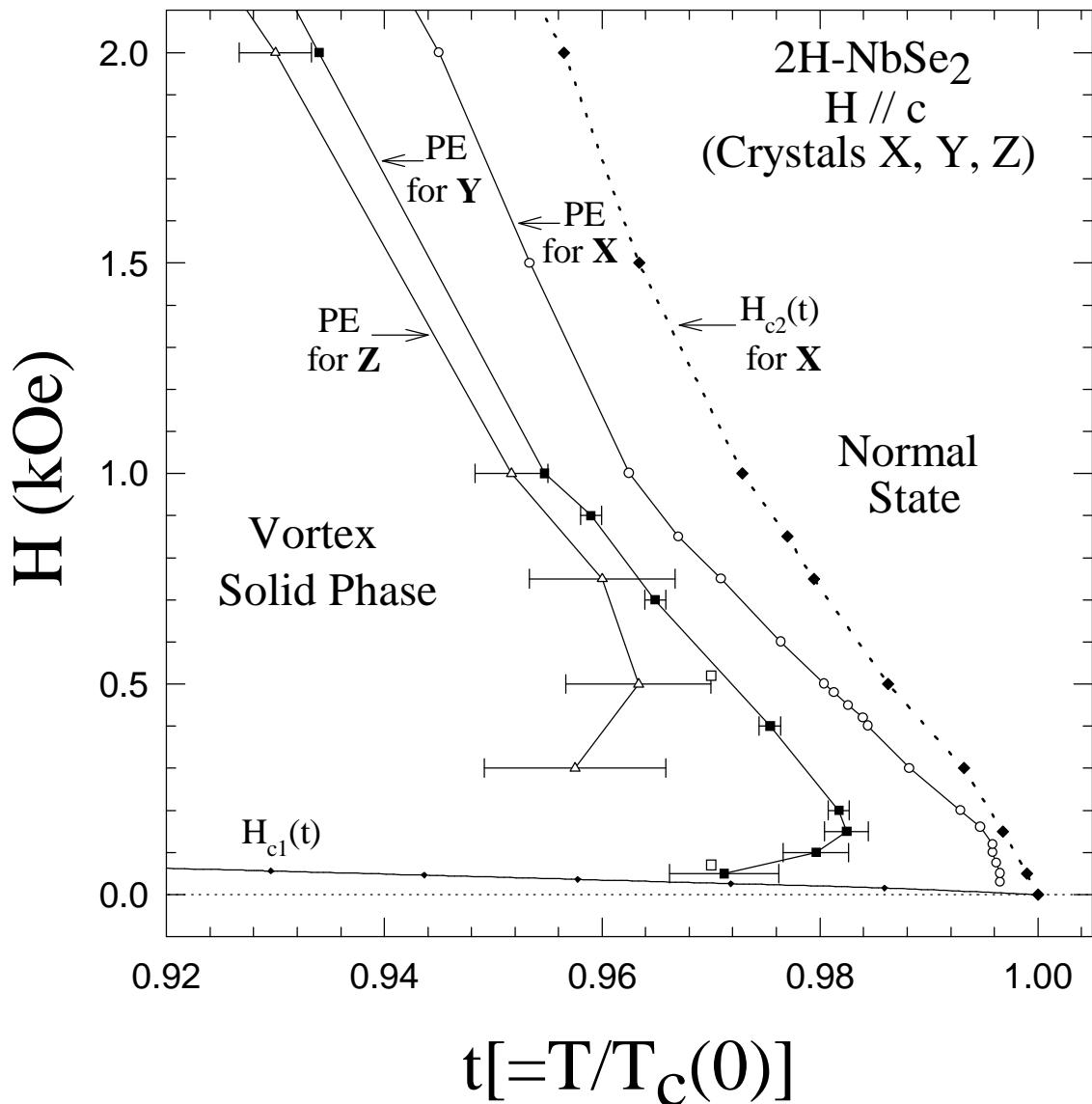


Fig. 4 (S. S. Banerjee)